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Atmospheric Chemistry and Physics

An unusual stratospheric ozone decrease in the Southern Hemisphere subtropics linked to isentropic air-mass transport as observed over Irene (25.5° S, 28.1° E) in mid-May 2002

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Abstract. A prominent ozone minimum of less than 240 Dobson Units (DU) was observed over Irene (25.5° S, 28.1° E), a subtropical site in the Southern Hemisphere, by the Total Ozone Mapping Spectrometer (TOMS) during May 2002 with an extremely low ozone value of less than 219 DU recorded on 12 May, as compared to the climatological mean value of 249 DU for May between 1999 and 2005. In this study, the vertical structure of this ozone minimum is examined using ozonesonde measurements performed over Irene on 15 May 2002, when the total ozone (as given by TOMS) was about 226 DU. It is shown that this ozone minimum is of Antarctic polar origin with a low-ozone layer in the middle stratosphere above 625 K (where the climatological ozone gradient points equatorward), and is of tropical origin with a low-ozone layer in the lower stratosphere between the 400-K and 450-K isentropic levels (where the climatological ozone gradient is reversed). The upper and lower depleted parts of the ozonesonde profile for 15 May are then respectively attributed to equatorward and poleward transport of low-ozone air toward the subtropics in the Southern Hemisphere. The tropical air moving over Irene and the polar one passing over the same area associated with enhanced planetary-wave activity are successfully simulated using the high-resolution advection contour model of Ertel's potential vorticity MIMOSA. The unusual distribution of ozone over Irene during May 2002 in the middle stratosphere is connected to the anomalously pre-conditioned structure of the polar vortex at that time of the year. The winter stratospheric wave driving leading to the ozone minimum is investigated by means of the Eliassen-Palm flux computed from the European Center for Medium-range Weather Forecasts (ECMWF) ERA40 reanalyses.

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1 Introduction

Tropical stratospheric ozone is a prominent actor in atmospheric chemistry and physics, and the southern tropical and subtropical latitudes are among locations where a possible recovery of the ozone layer may be detected. However, tropical ozone studies that rely only on satellite measurements are not able to neither resolve vertical-fine scale structures nor completely improve our understanding of both photochemical and dynamic processes that are operating in the atmosphere and contributing to the ozone budget. The sparseness of in-situ measurements in the tropical and subtropical Southern Hemisphere has limited investigations of ozone distribution and variability related to atmospheric dynamics and climate, e.g., the meridional transport, the varying position of the Intertropical Convergence Zone (ITCZ), the Quasi-Biennial Oscillation (QBO), the El Niño-Southern Oscillation (ENSO) and La Niña.

Situated in the subtropical region of the Southern Hemisphere, Irene represents a location of major interest for the observation of low and high latitude influences attributed to transport processes. Thompson et al. (2003a, b) used 1100 SHADOZ radiosondes from 10 southern tropical and subtropical sites during the 1998–2000 period to characterize the seasonality and variability in ozone. They showed that the total amount of ozone is generally low in the tropics in winter. Their data also show higher stratospheric ozone at Irene due to a greater frequency of mid-latitude air passing over the site

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in the lower stratosphere. Furthermore, recent observational studies have shown that in the southern subtropics, within the UTLS region, many dynamical processes, such as subtropical tropopause breaks (Baray et al., 1998), and isentropic horizontal exchanges in the low stratosphere through the southern subtropical barrier (Bencherif et al., 2003; Portafaix et al., 2003) take place. Using a middle atmosphere circulation model, Horinouchi et al. (2000) showed that the transport between the tropics and the extratropics is strongly dependent on altitude and has geographic preferences in the lower stratosphere. Calisesi et al. (2001) stressed the role of southward excursions of the polar vortex in the large episodic winter ozone perturbations observed in the mid-latitude middle stratosphere. Additionally, Koch et al. (2002, 2005) showed that the extreme total ozone anomalies in the mid-latitude stratosphere are associated with meridional transport from regions with climatologically low/high ozone concentrations. Riese et al. (2001) studied transport of tropical air southward in association with a strong interaction of the South polar vortex with the subtropical barrier.

The present paper reports on an unusual stratospheric ozone decrease recorded during May 2002 over Irene and highlights its relationship with the early winter phase of the unusual Southern Hemisphere winter 2002 in which the first major sudden stratospheric warming (SSW) was detected since regular monitoring began in the 1940s (Krüger et al., 2005). The exceptional SSW was followed by a split of the ozone hole into two parts in September 2002 (World Meteorological Organization (WMO), 2003). In fact, an unusually weak polar vortex was an exceptional feature of the entire winter 2002 in the Southern Hemisphere, which preconditioned it for a progressive dilation. This was associated with distinctive persistent stratospheric vacillations starting around 20 June 2002 (Scaife et al., 2005). This behavior of the wintertime polar vortex was considered to be the main perquisite for the SSW of September 2002. This latter unprecedented event has been extensively analyzed (e.g., Journal of the Atmospheric Sciences (JAS) Special Issue, 2005). Nevertheless, the 2002 early-winter pre-conditioning anomalies and their impact on the subtropics have been little documented and studied.

Here the high-resolution advection model MIMOSA (Modélisation Isentrope du transport Méso-échelle de l'Ozone Stratosphérique par Advection), and the Eliassen-Palm flux computed from the European Center for Medium-range Weather Forecasts (ECMWF) ERA40 re-analyses, are used to investigate the basic dynamics behind the aforementioned ozone decrease event observed over Irene in Mid-May 2002.

The data and analytical tools used in this study are described in Sect. 2. In Sect. 3, we characterize the May 2002 ozone anomaly using 7 years of Total Ozone Mapping Spectrometer (TOMS) and ozonesonde data. The dynamical processes are investigated in Sect. 4. Conclusions are presented in Sect. 5.

2 Data and analysis

In this section, a brief overview of the ozone and meteorological data along with the diagnostic tools used for the analyses is given.

2.1 Data description

2.1.1 Ozone data

The 1998-2005 ozone profiles for Irene (25.5° S, 28.1° E, Pretoria, South Africa) used in this study have been performed by the SAWS (South African Weather Service) and are archived on the SHADOZ (Southern Hemisphere Additional Ozonesondes) web site (http://croc.gsfc.nasa.gov/shadoz) (Thompson et al., 2003a, b). The ozonesonde used is of Science Pump ECC6A type with 1% KI buffered solution, and the meteorological sonde is of Väisälä RS80-15GE type. For the present study, 178 profiles measured fortnightly between November 1998 and May 2005 were used. More precisely, in order to examine the early winter state of stratospheric ozone over Irene, we focused our analysis on the May ozone concentration profiles measured during the period from 1999 to 2005. In addition, total ozone columns over Irene for the same period were taken from the TOMS experiment on board the Earth Probe satellite (Earth Probe TOMS V.8 overpass data available at NASA/Goddard Space Flight Center web site: http://toms.gsfc.nasa.gov/ozone), which provides daily global distribution of ozone with a resolution of 1° in latitude and 1.25° in longitude.

2.1.2 Meteorological data

Information about the dynamical evolution of the atmosphere in the early southern winter 2002 is provided by the ERA-40 re-analyses archived on the ECMWF web site (http://data.ecmwf.int/data/d/era40_daily/). In this study, ECMWF horizontal winds and temperature were extracted on a 2.5°×2.5° grid from 1000 to 1 hPa (23 levels) at 00:00, 06:00, 12:00 and 18:00 UT, for the period extending from 1 April to 31 May 2002.

2.2 Diagnosis tools

2.2.1 The Ertel's potential vorticity and the Eliassen-Palm flux

In this paper, the dynamical processes are investigated using the following diagnostic tools computed from the ECMWF data: the isentropic Ertel's potential vorticity (Epv) and the Eliassen-Palm (E-P) flux.

First, the Epv on isentropic surfaces, which behaves as a dynamical tracer in the absence of diabatic effects, is used to study the isentropic transport across the dynamical barriers in the stratosphere: polar vortices or subtropical barriers (Hoskins et al., 1985; Holton et al., 1995; Bencherif et al.,

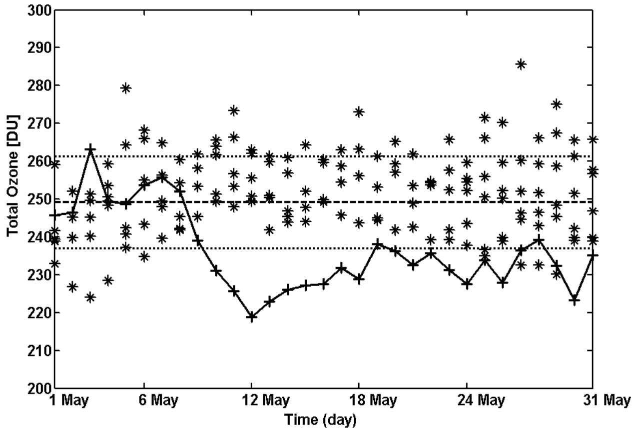

Fig. 1. Daily total ozone column (in DU) for May, over Irene, as derived from TOMS/Earth Probe Satellite overpass data for 2002 (solid line with "+" symbols) and for 1999–2005 ("*"). The horizontal lines represent the corresponding monthly mean value (dashed line) $\pm\sigma$ (dotted line).

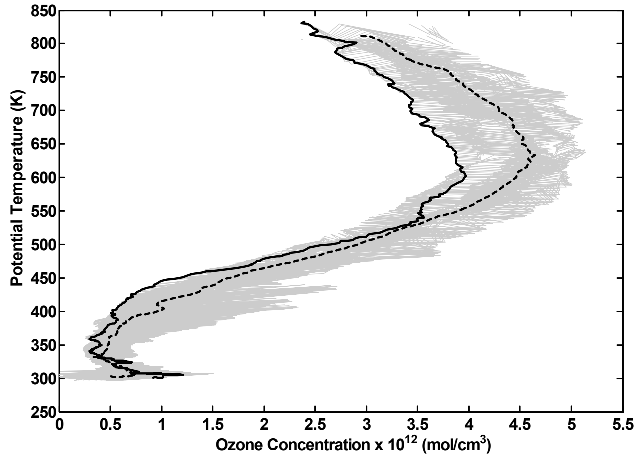

Fig. 2. Ozone concentration profile as obtained from ozonesonde measurements over Irene (25.5° S, 28.1° E) on 15 May 2002 (solid line) compared to the May profiles from 1999 to 2005 (in grey) and the corresponding mean profile (dashed line).

2003). Here the Epv is evaluated from ECMWF horizontal wind and temperature fields interpolated onto isentropic surfaces.

Second, the E-P flux vector ($\boldsymbol{F}$) and its divergence (div($\boldsymbol{F}$)) are used to obtain information about the planetary-wave activity and the possible wave-breaking regimes. The orientation of the E-P flux vector indicates the direction of planetary-wave propagation (Andrews et al., 1987). Planetary-wave activity in the mid-latitudes generally propagates from the winter troposphere up into the stratosphere and mesosphere and toward the equator (Eliassen and Palm, 1961; Kanzawa et al., 1984). Planetary-wave breaking can be recognized by the convergence of the E-P flux vectors (div($\boldsymbol{F}$)<0).

2.2.2 The MIMOSA advection transport model

We use the MIMOSA model in order to further investigate the isentropic transport across the dynamical barriers in the stratosphere, and to evaluate the contribution of this (horizontal) transport mechanism in the vertical distribution of ozone over the subtropics. MIMOSA is a high-resolution advection contour model of Ertel's potential vorticity, which was developed at the Service d'Aéronomie by Hauchecorne et al. (2002). The model runs on an orthogonal grid covering the whole Southern Hemisphere with a resolution of 3 grid points/degree. The Epv at each grid point is advected with a time step of 1 h using ECMWF ERA-40 wind fields and the advected Epv (denoted APV hereafter) fields are re-interpolated onto the original grid every 6 h. The diabatic evolution of the Epv field at large scales is extracted from ECMWF fields by applying a relaxation toward the ECMWF Epv field with a time constant of 10 days (Morel et al., 2005). In the present study, the MIMOSA model is run for two months starting on 1 April 2002.

3 Ozone observations over Irene in May 2002

This section is designed to characterize an extreme ozone event in the stratosphere observed during May 2002 over Irene. The specific date of May 15 of our study is chosen by matching particularly low ozone events identified from Earth Probe TOMS records with a coinciding ozonesonde flight over Irene.

Daily total ozone values derived from TOMS records for years between 1999 and 2005 are depicted for the month of May in Fig. 1. As shown by the dashed horizontal line on the figure, the monthly averaged total ozone over Irene location is 249±12 DU (at 1σ). The absolute minimum total ozone (219 DU), which is about 30 DU less than the May climatological mean, is obtained on 12 May 2002. As for the coincident day of ozonesonde flight (on 15 May), the corresponding total ozone (226 DU) is also significantly less than the climatological mean. In fact, one notices that the negative anomaly of total ozone persists for more than a week (see Fig. 1).

The mid-May vertical distribution of stratospheric ozone over Irene as obtained from ozonesonde measurements is illustrated in Fig. 2. It shows the ozone concentration profile (solid line) recorded on 15 May together with the monthly mean profile (dashed line). The later is obtained similarly as for the TOMS total ozone mean, i.e. by averaging together all the May ozone profiles (over 16 ozonesondes flown fortnightly from 1999 to 2005).

The ozone profile recorded on 15 May 2002 (Fig. 2, solid line) shows strong negative deviations in comparison with

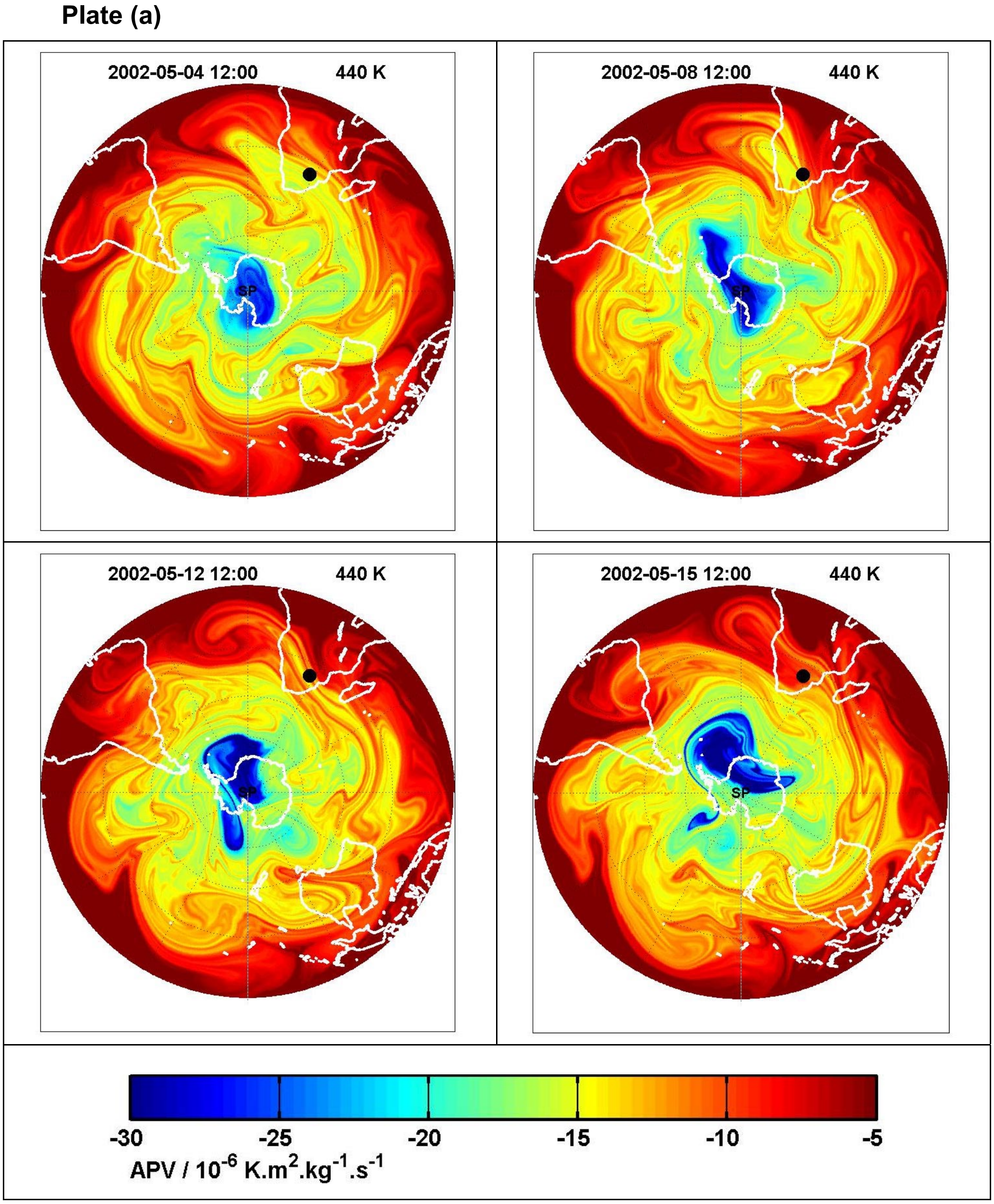

Fig. 3. Snapshots of isentropic Ertel's potential vorticity advected by the MIMOSA advection transport model. Plate **(a)** Outputs are calculated on the 440-K isentropic level for 4, 8, 12 and 15 May 2002, Plate **(b)** Outputs are calculated on the 675-K isentropic level for 4, 8, 12 and 15 May 2002 and Plate **(c)** Top panel: Temporal evolution of advected potential vorticity (APV) as a function of potential temperature obtained from the high-resolution MIMOSA model at Irene in May 2002. Bottom panel: same as top panel but zoomed in on the vertical range from 380 to 520 K. Irene is indicated on APV-maps with a black spot.

the 7-year (1999–2005) mean profile for May (dashed line) between 400-K and 450-K in the lower stratosphere and above the 625-K potential temperature level in the middle stratosphere. This suggests that the total ozone decrease reported from TOMS data in the early winter of 2002 (mid-May) and depicted in Fig. 1 may be related to the (very) low concentrations of ozone at isentropic levels between 400-K and 450-K and at those greater than 625 K (Fig. 2).

4 Isentropic transport and the mid-May 2002 ozone minimum

The aim of this section is to investigate the role of isentropic transport of tropical and polar air masses, in conjunction with an increase in planetary-wave activity and the induced isentropic mixing, to contribute to the extreme ozone reduction event observed in early winter 2002 in the subtropics (Irene).

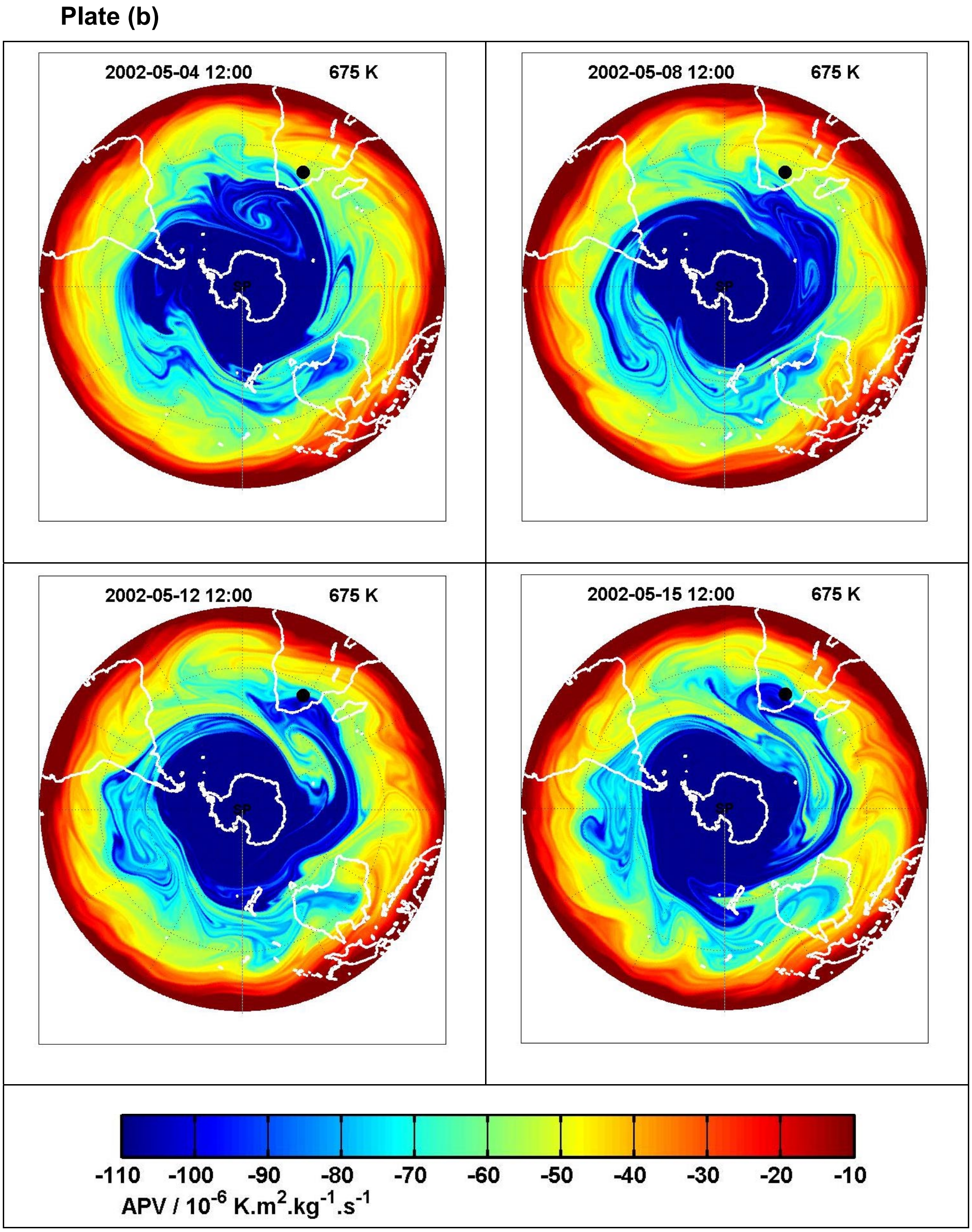

Fig. 3. Continued.

4.1 Tropical and polar air mass advection toward the subtropics

In order to investigate tropical and polar air-mass transports toward the subtropics, high resolution PV-maps on selected isentropic surfaces were constructed for 4–15 May using the MIMOSA model.

Plate (a) and (b) of Fig. 3 show snapshots of Epv advected by MIMOSA (APV) on the 440-K (lower stratosphere) and on the 675-K (middle stratosphere) isentropic surfaces for selected days prior to and during the ozone minimum event. The tropical and polar air masses can be identified respectively by low and high absolute APV values. On each APV-map, the location of Irene is indicated by a black spot. On 4 and 8 May (upper APV maps on plate (b) of Fig. 3), at the 675-K isentropic surface, Irene is covered by air-masses of relatively low absolute APV values, while on 12 and 15 May, a tongue with high absolute APV indicating air of polar

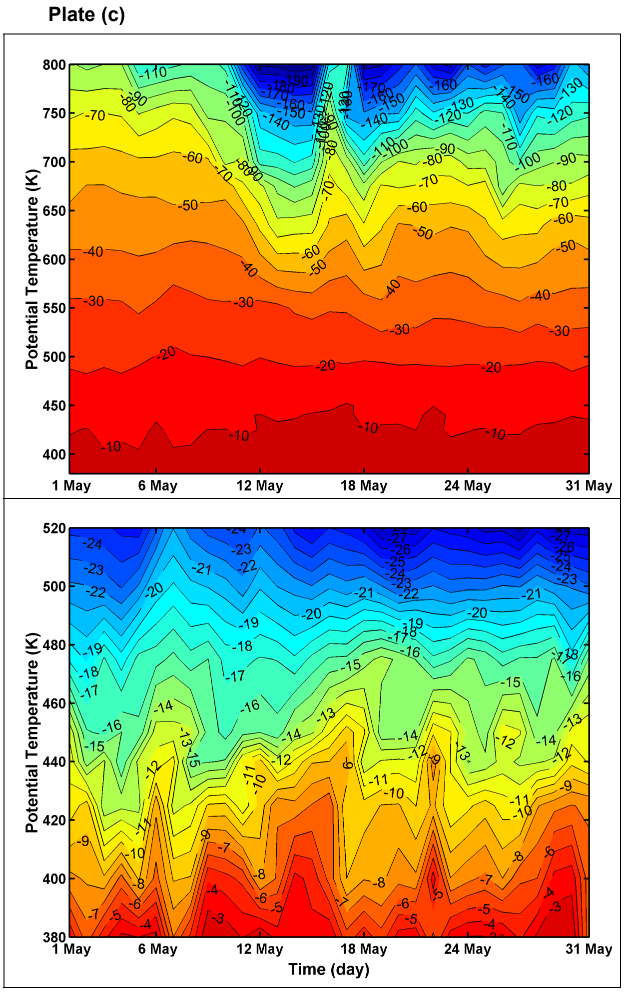

Fig. 3. Continued.

origin is deformed and shifted away from the pole toward the subtropics. It extends over the 15–120° E longitude and 20–40° S latitude range, covering a large area over the south part of Africa, including Irene. In parallel, on 4 and 8 May (upper APV maps on plate (a) of Fig. 3), at the 440-K isentropic surface, Irene is covered by air-masses of relatively high absolute APV values, while on 12 and 15 May, a tongue with low absolute APV indicating air of tropical origin, has moved eastward and southward toward the subtropics.

Nearly the same transport situations are obtained from MIMOSA outputs for selected isentropic surfaces in the 625–800 K range (not shown). Furthermore, Plate (c) of Fig. 3 shows the time evolution of APV as a function of potential temperature over Irene during May 2002 in the stratosphere. During the mid-May period, the Irene site is under the influence of polar air masses in the middle stratosphere (top panel) and of tropical ones in the lower stratosphere (bottom panel). This is in agreement with the vertical extension of the negative deviation observed on the ozone concentration profile recorded on 15 May for isentropic levels higher than 625 K and for those between 400 K and 450 K (see Fig. 2).

Because of polar vortex disturbances, MIMOSA analyses show how polar air masses were injected into mid-latitude regions and sporadically into the subtropics. Moreover, this large latitudinal extension (from pole to subtropics) goes simultaneously, in a reverse way, with isentropic transport of tropical air masses towards the mid-latitudes in the lower stratosphere. This episode of horizontal exchange between the tropical stratospheric reservoir and mid-latitudes is also well identified on MIMOSA APV-maps during the period from 10–18 May (not shown). This is in agreement with the time extension of the minimum of total ozone derived from TOMS measurements (see Fig. 1).

Thus, the unusual reduction of total ozone observed over Irene by mid-May 2002 seems to be related to isentropic transport of air masses simultaneously in the lower and middle stratosphere, respectively from the tropics to the mid-latitudes and from the pole to the subtropics.

It is a particularly interesting situation. In the lower stratosphere (400–450 K) the ozone profile (Fig. 2) shows a tropical influence. Indeed, ozone concentrations there are significantly below climatological values and similar to the tropical values in the lower stratosphere (Portafaix et al., 2003). In the middle stratosphere, the low concentrations of ozone both in the upper part of the profile (above 625 K) and on the MIMOSA APV maps can be attributed to air-mass advection from pole to tropics as the climatological ozone decreases towards high latitudes in the middle stratosphere of both hemispheres (Koch et al., 2002; Godin et al., 2002). The mid-stratospheric ozone decrease observed over Irene appears hence to be a local effect of the large-scale disturbances occasionally affecting the polar stratospheric circulation as early as May 2002. Given its relationship to these large-scale features and their transient character, this mid-stratospheric ozone decrease is of purely dynamical origin.

4.2 Wave activity

A perturbed polar vortex is associated with enhanced planetary-wave activity, which contributes to pull out material from the vortex and distribute filaments equatorward (Schoeberl et al., 1988, 1992). Moreover, in the stratosphere nearby a subtropical barrier the isentropic mixing has been linked to disturbances occurring at the vicinity of the polar vortex in the winter hemisphere (Waugh, 1993). According to Marchand et al. (2003), the transport of polar air toward midlatitudes can occur through the extension of filament toward lower latitudes and through vortex intrusions. This transport can have different effects depending on whether it is reversible or irreversible. If reversible, the effect is to perturb the ozone content at low latitudes for a limited period of time. If irreversible, polar air is mixed with the surrounding air.

The rapid and irreversible deformation of Epv contours on the 675-K isentropic surface observed in plate (b) of Fig. 3 suggests a planetary-wave breaking linkage resulting

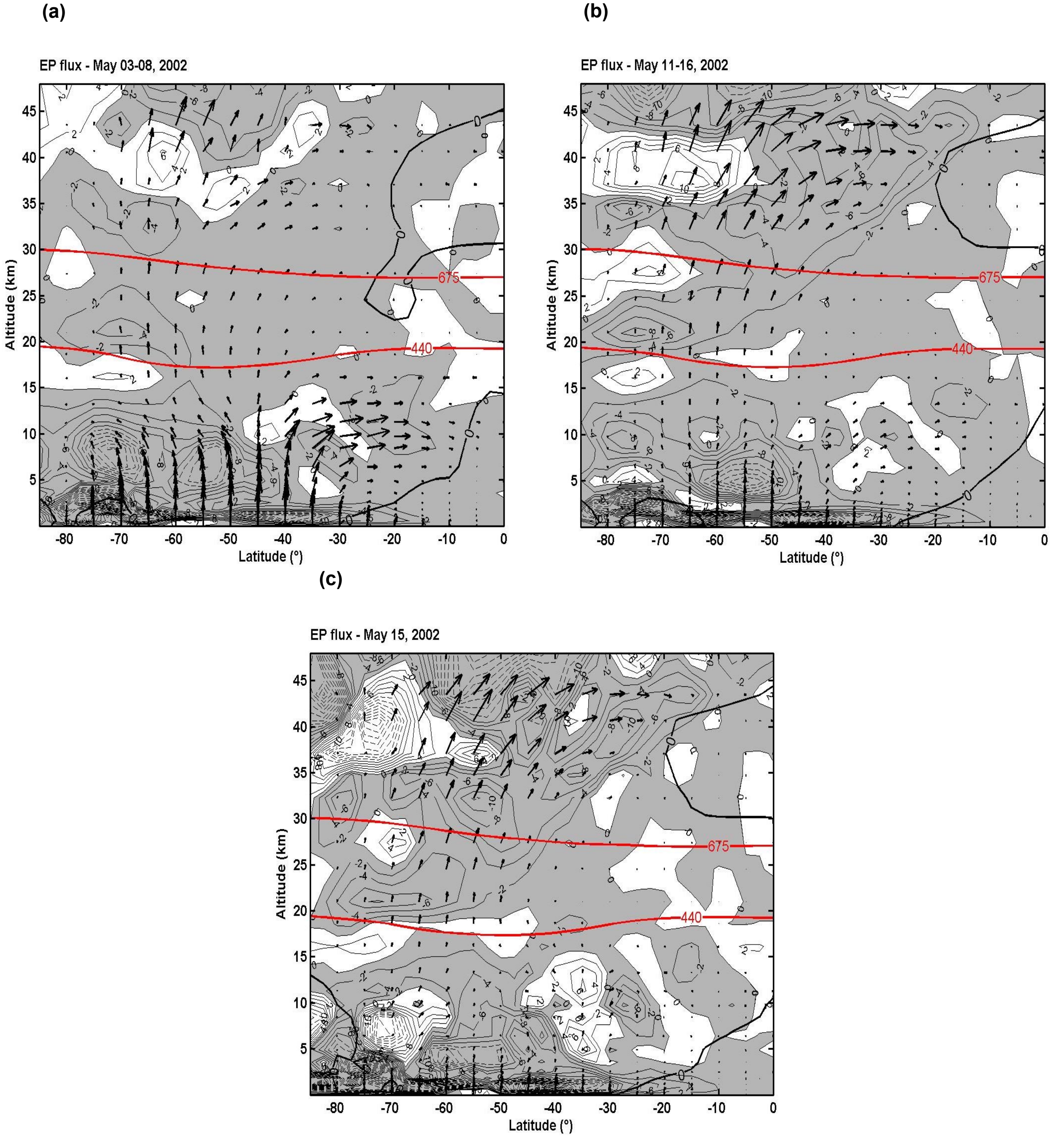

Fig. 4. E-P flux (***F***) cross-sections in the meridional plane averaged over **(a)** 3–8 May, **(b)** 11–16 May and **(c)** for the selected day of 15 May 2002. Contours represent div (***F***), in m.s^{-1} per day; negative values are shaded. The contour interval is 2 m.s^{-1} per day and dashed contours correspond to values more than 10 m.s^{-1} per day or less than -10 m.s^{-1} per day. The zero wind line has been overlaid (thick solid contour). The red lines indicate the isentropes 440-K and 675-K.

in quasi-horizontal mixing and irreversible tracer transport (McIntyre and Palmer, 1983, 1984).

Figure 4 shows E-P cross-sections computed using ECMWF ERA40 re-analyses; with arrows representing the E-P flux vectors (***F***), and contours represent div (***F***), averaged over the period 3–8 May (Fig. 4a) and 11-16 May

(Fig. 4b) and for the selected date of 15 May 2002 (Fig. 4c). Planetary-wave breaking is identified by the convergence of the E-P flux (i.e. negative values in Fig. 4). Figure 4a shows enhanced wave activity in the troposphere for 3–8 May, which has led to the increase of wave activity later in the stratosphere. Indeed, the comparison between Figs. 4a and 4b shows an increase in stratospheric wave activity during the period from 11 to 16 May. In fact, strong upward wave propagation is located over high latitudes during that period of time (Fig. 4b). The E-P flux vectors bend equatorward with height and generate a large region of convergence over the subtropics in the stratosphere, where the wave driving reaches a minimum lower than $-6\,\mathrm{m.s^{-1}}$. The wave activity is particularly strong on 15 May 2002 (Fig. 4c) and is associated with a greater wave penetration and an enhanced wave driving in the subtropical upper stratosphere where the wave driving reaches a minimum lower than $-10\,\mathrm{m.s^{-1}}$ per day. This analysis demonstrates that, by early-winter 2002, planetary-wave activity has significantly increased during the mid-May period. It shows upward and equatorward planetary-wave propagation.

The southern stratospheric zonal circulation changed from easterlies to westerlies during the early winter of 2002, allowing the planetary waves to spread and bend equatorward nearby the subtropics (as shown by EP-flux on Fig. 4). According to Newman and Nash (2005), the subtropical zonal wind in the upper stratosphere in April 2002 (prior to any wave events) was anomalously easterly. Therefore, the anomalous easterlies in the mid-to-upper stratosphere in April corroborated by a particular reversal to westerlies in early May, suggests that wave propagation was highly anomalous during the early winter of 2002. The mechanism for this appears to be the dependence of planetary wave propagation and breaking on the structure of the mean zonal wind (Brasseur et al., 1999). In fact, when the subtropical wind is westerly, waves can propagate as far as toward the equator, and the surf zone is shifted to lower latitudes. Thus, the two anomalous features both in the zonal wind and in the planetary-wave activity contribute to the increase in isentropic mixing over the subtropics by mid-May 2002. This is a result of mixing by planetary wave breaking in the surf zone.

Clearly, the early-winter dynamics of 2002 is directly responsible for the unusual ozone reduction observed over Irene in mid-May 2002. The large-scale transport and mixing of polar air-masses explains the decrease of stratospheric ozone over Irene and the strong negative deviations recorded in mid-May 2002 in Irene ozone profile when compared with the 7-year (1999–2005) mean May profile. Indeed, the polar vortex in the early winter of 2002 was unusually disturbed so that enhanced planetary-wave activity easily eroded it into filaments. This gave rise to large-scale transport of polar air toward the subtropics in the middle stratosphere and contributed to the development of the low ozone episode of a purely dynamical origin over Irene in mid-May 2002.

5 Discussion and conclusion

In this paper, we investigated the ozonesonde dataset obtained at Irene, a South-African subtropical site operating under the SHADOZ project. The retrieved ozone concentration profiles were supplemented by daily TOMS total ozone columns derived for the same location and covering the same period, i.e., November 1998–May 2005.

A prominent ozone minimum has been reported in mid-May 2002 from the TOMS and ozonesonde datasets. The combination of these datasets suggests that the most significant contribution to the total ozone reduction is of dynamical origin and may be explained by low ozone concentrations obtained (1) at isentropic surfaces higher than 625 K in the middle stratosphere and (2) at those between 400 K and 450 K in the lower stratosphere. The absolute minimum of total O_3 (219 DU) was 30 DU (about 12%) less than the mean value for May.

It was found from MIMOSA APV-maps that the observed ozone reduction over the subtropics (Irene) could be attributed to the transport of tropical and polar air masses. The reversal to westerlies in the subtropics allows the planetary waves (PWs) to propagate further to the equator, so that they can break at the subtropical barrier. The propagation of the PWs can be seen in APV-maps in the middle stratosphere (polar tongue), whereas the breaking of PWs (see div ($\boldsymbol{F}$) in Fig. 4) leads to erosion of the subtropical barrier.

The present study demonstrated that the early-winter dynamics of 2002 was responsible for the unusual ozone reduction over the subtropics through large-scale transport and mixing of tropical and polar air masses. In fact, the early-winter 2002 ozone minimum and its large extension up to the subtropics represent an anomaly, which is connected to the unprecedented state of the southern polar vortex disturbances recorded during May 2002 and reported by Newman and Nash (2005).

To summarize, a 8–12% decrease in total column of ozone, concomitant with low-ozone concentrations in the middle stratosphere at isentropic levels above 625 K and in the lower stratosphere (400–450 K) observed over Irene in mid-May 2002, is of dynamical origin. Indeed, it can be attributed respectively to ozone-poor air originally from the polar vortex and to ozone-poor air coming from tropics. This resulted in the lowest ozone column recorded during the 7-years period (1998–2005). MIMOSA APV-maps representing the early winter 2002 period in the middle stratosphere highlighted an unusually high planetary-wave activity and a disturbed polar vortex with filament excursions and strong mixing up to the subtropics in the middle stratosphere. In parallel, MIMOSA model simulated successfully the transport of a tropical poor-ozone air toward the subtropics in the lower stratosphere.

The exceptional presence of polar vortex air over the subtropics during May 2002 can be considered as the first sign of the particular polar vortex disturbances, which after being well reinforced, contributed to the unprecedented behavior of the Antarctic spring ozone hole observed during September 2002.

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